Li-Ion Cell Manufacturing Using Directly Recycled Active Materials

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DOE VTO Annual Merit Review

Project ID: bat356

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Overview

Timeline

- Start: February 2017
- End: January 2019
- Percent complete: 66 %

Budget

- $1.8M total project budget:
  - $900k DOE
  - $900k Farasis
- 50 % Cost share

Barriers

- Cost/value recovery of current battery recycling processes
- Quality of feedstock for direct recycling
- Complexity and variability of LiB designs

Partners

- Farasis Energy, Inc
- Lawrence Berkeley National Laboratory – R. Kostecki group

Relevance / Objectives

- **Project Goal:** The goal of this project is to develop recycling technology for Li-ion batteries that will enable direct reuse of valuable active materials.
- **Performance Objective:** The objective is to demonstrate the utility of direct recycling technology by producing cells with recycled active materials that have performance within 5% of control cells using pristine versions of the same active materials.
  - Optimized recovered materials and formulations will be used to manufacture large pouch cells (25 Ah) as project deliverables.

Project Milestones

Tasks	Milestone		Project Month	Status
Task 1	1.3.1	Final Report summarizing initial electrochemical testing	24	on track
Task 2	2.1.1	Acquisition of direct recycling process equipment	3	**Complete**
	2.2.1	Completed installation of direct recycling pilot line	5	**Complete**
	2.3.1	Recovery of 2 kg Positive AM & 1 kg Negative AM from manufacturing residues	8	75 % Complete
	2.3.2	Recovery of 2 kg Positive AM & 1 kg Negative AM from EOL cells	14	*delayed 2 mo.*
	2.3.3	Recovery of 2 kg Positive AM & 1 kg Negative AM from EV battery modules	17	on track
Task 3	3.1.1	Demonstrate density based separation at a scale of 5 kg black mass input	10	*delayed 3 mo.*
	3.1.2	Improve separation yield to >95%	17	on track
	3.1.3	Recover direct recycled active materials in greater than 99.9% purity.	17	on track
	3.2.1	Demonstrate recovered active materials with specific capacities and first cycle efficiencies identical to pristine materials	17	on track
	3.2.2	Assessment of economic impact of surface area reduction processing	18	on track
	3.2.3	Report on detailed materials characterization of recycled active materials.	20	on track
Task 4	4.1.1	Demonstrate separation of mixed spinel/layered oxide cathode material mixtures using density-based separation.	19	on track
	4.2.1	Demonstrate reconditioning of mixed spinel/layered oxide cathode material mixtures	19	on track
Task 5	5.1.1	Completion of Cell Build 1	12	*delayed 5 mo.*
	5.2.1	Completion of Cell Build 2	16	*delayed 5 mo.*
	5.3.1	Completion of Deliverable Cell Build	21	on track
	5.3.2	Delivery of controls and cells with > 50% recycled active material content.	22	
Task 6	6.1.1	Delivery of initial test data	24	on track
	6.2.1	Quantification of impact of recycled active materials on technology lifetime and cost	24	on track

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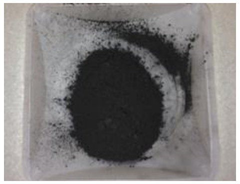
Approach – Direct Recycling Process Overview

Discharged cells

Shredding

Electrolyte extraction

Sieving



Black Mass

Density Separation

Graphite

$LiMO_x$

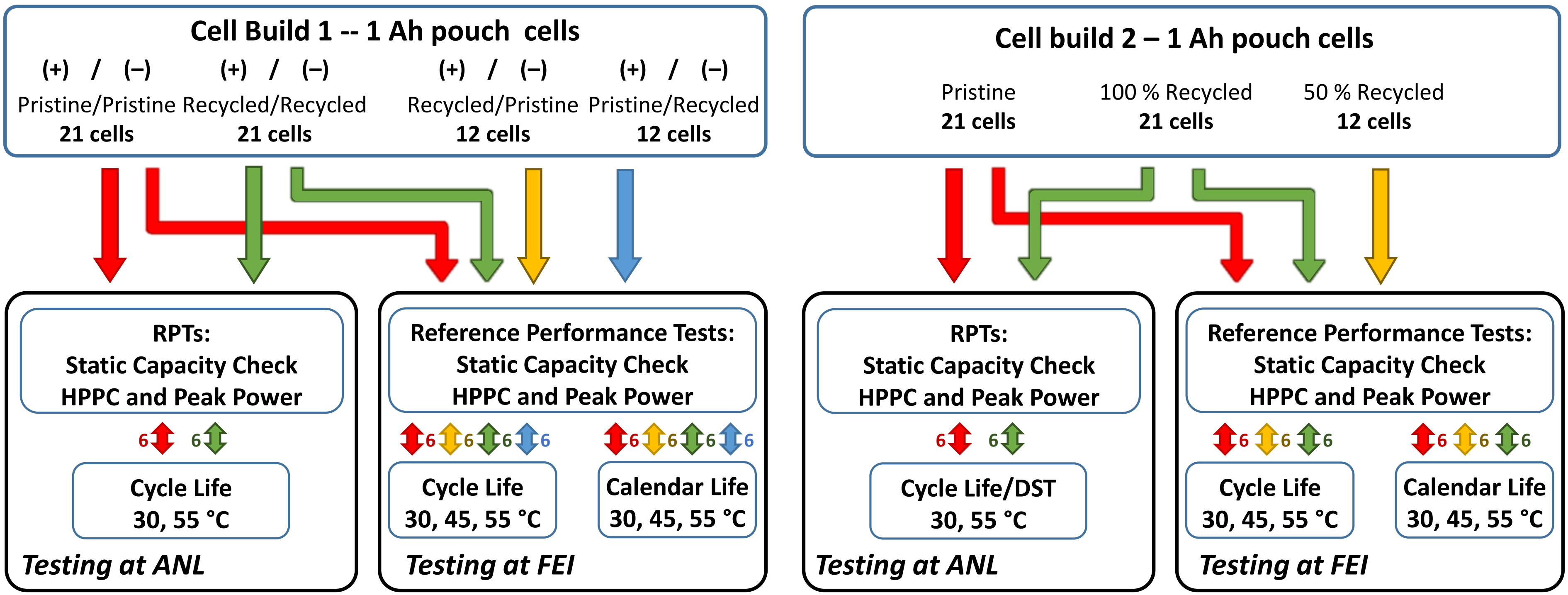
Purification

Regen-eration

Recycled Materials

- Direct recycling uses only physical separation processes
- Active materials are recovered essentially intact, thus capturing some of the value added during original material synthesis
- Chemical purification and re-lithiation are performed under relatively mild conditions with low energy intensity

Approach – Cell Builds



- Intermediate cell build 1 examines different combinations of recycled and pristine active material electrodes to test for cell performance sensitivity to either recycled active material.
- Intermediate cell build 2 provides additional data to optimize blending of recycled and pristine active materials for the final deliverables.

Approach - Complexity

Recycling Feedstocks

This project evaluates multiple possible inputs for direct recycling:

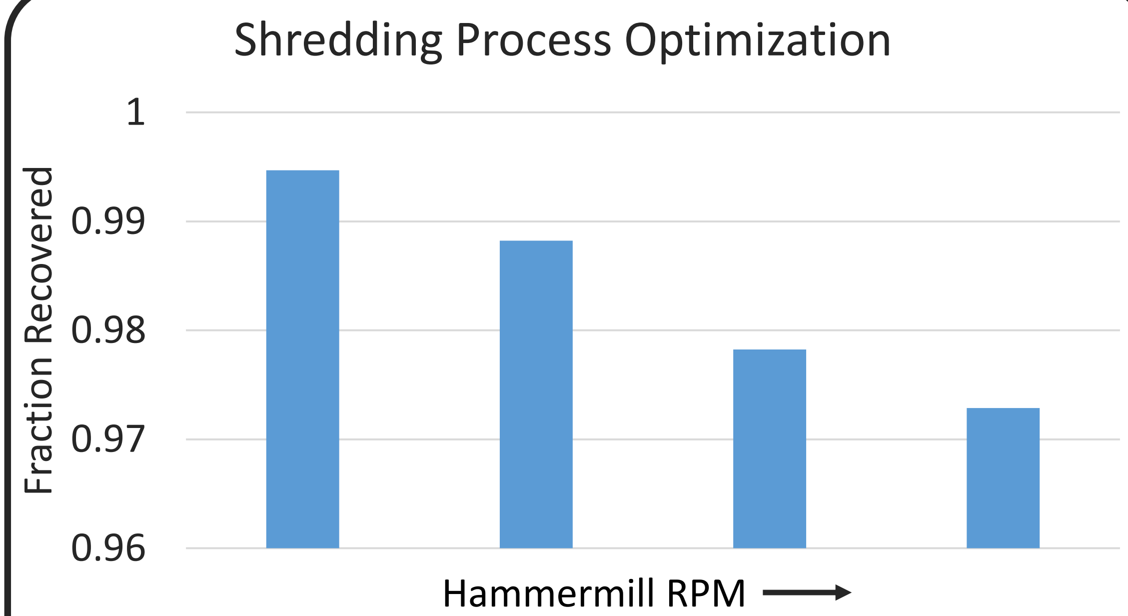
- **Electrode production scrap**
- **Formed cells**
- **Entire battery modules**

Cell Chemistries

- **First large-scale feedstock for commercial scale recycling NCM111 is the main focus for process development and deliverables**
- **Additional process development is being performed for more complex mixed active material cathodes: LMO+NCM**
- **Other cathode chemistries (NCM523, LFP, …) will be evaluated for compatibility with optimized processes**

Technical Accomplishments – Recycling Process Scale-up

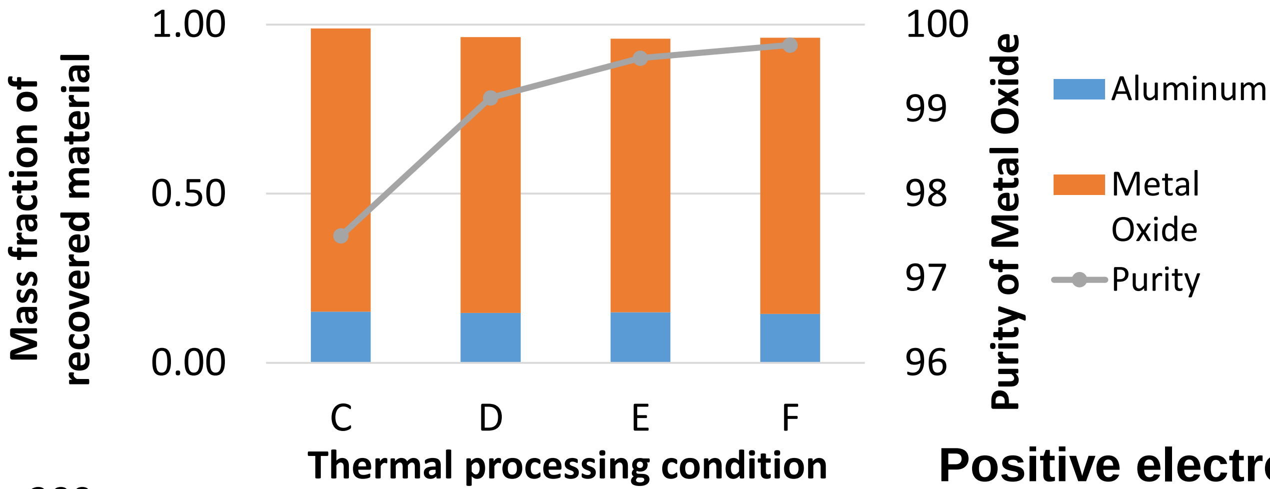
- Completed facility modifications for scaled-up recycling operations.
- Shredding equipment is housed in a large fume hood for airborne particulate control.
- Facility is capable of producing several kg per day of recycled active materials.

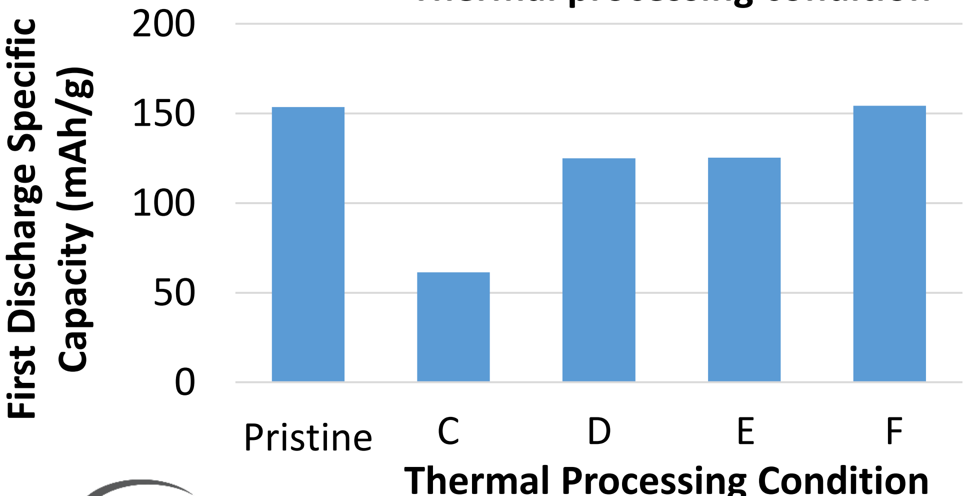


- Material recovery increases as milling power decreases due to less dust generation.

Technical Accomplishments – Positive Electrode Thermal Treatment

Single stage thermal treatment of shredded positive electrode scrap

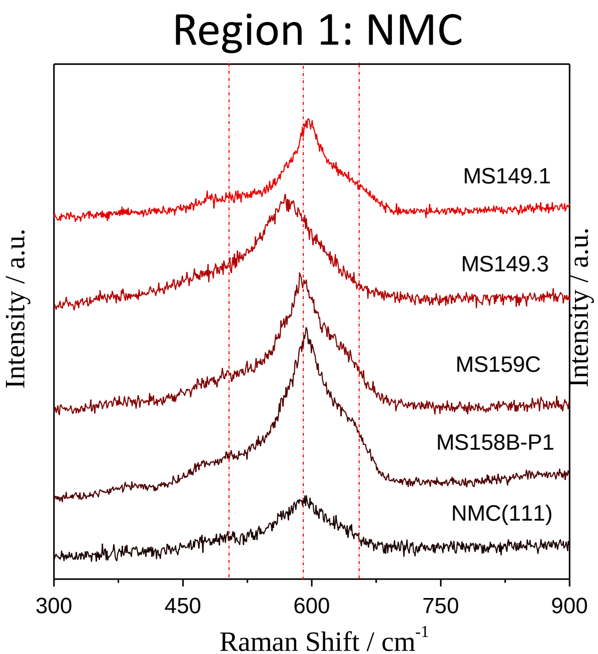


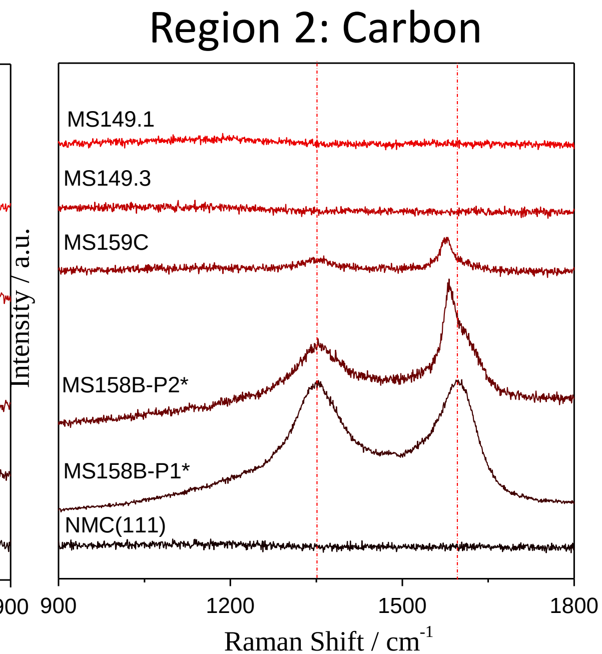


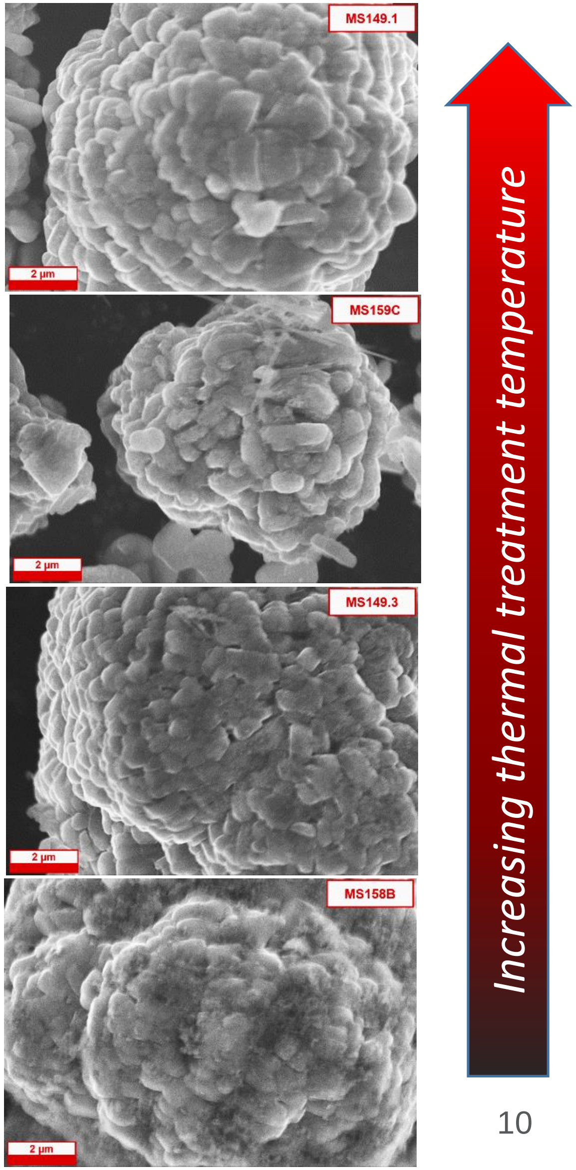
Positive electrode thermal treatment:

- Thermal pretreatment is performed to degrade binder and conductive carbons
- Purity of recovered metal oxide increases with increased processing temperature, but the m.p. of Al current collector limits temperature at this stage

Technical Accomplishments – Positive AM Characterization







- Raman spectrometry shows that conductive carbons are decomposed with sufficiently high thermal treatment temperature.
- SEM visually confirms cleaner particle surfaces with higher treatment temperatures.

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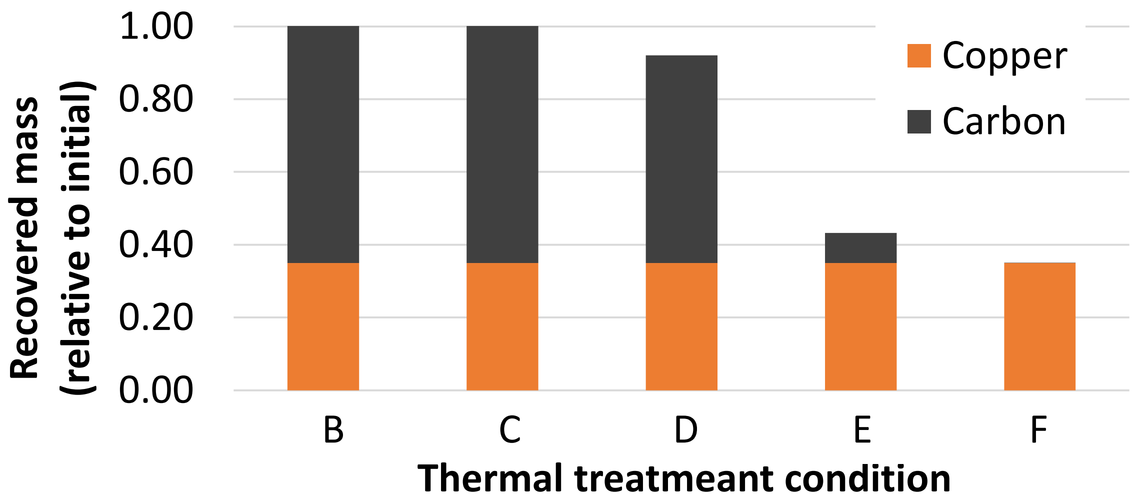
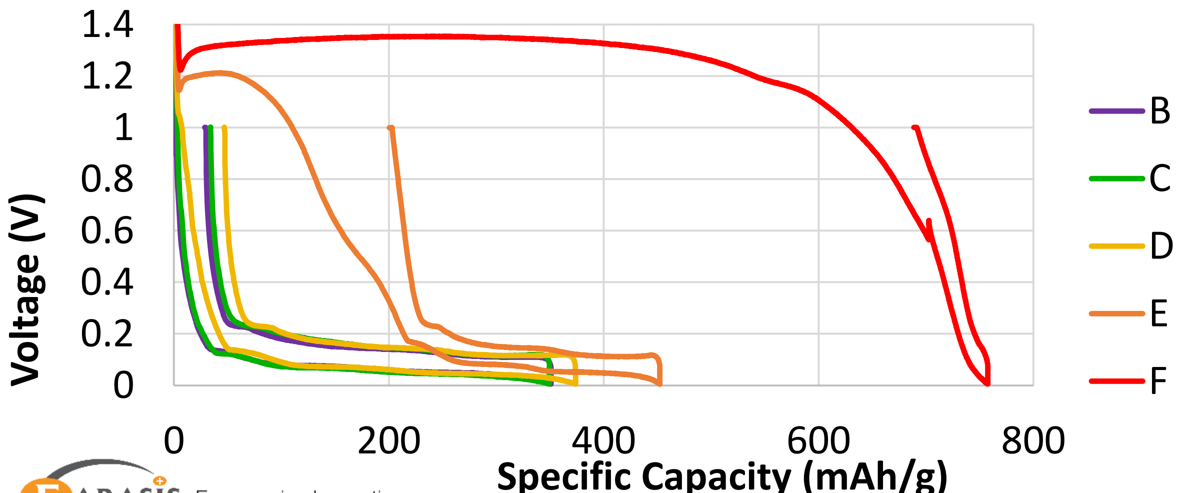
Technical Accomplishments – Negative Electrode Thermal Treatment

Single stage thermal treatment of shredded negative electrode scrap





- Increasing thermal exposure diminishes recovery of graphite. Mass loss is due to carbon combustion: C (*s*) + O_2 (*g*) → CO_2 (*g*).
- During thermal processing the copper foil is also oxidized to varying degrees. The lowest temperature conditions produce high yields of graphite with minimal Cu oxidation.

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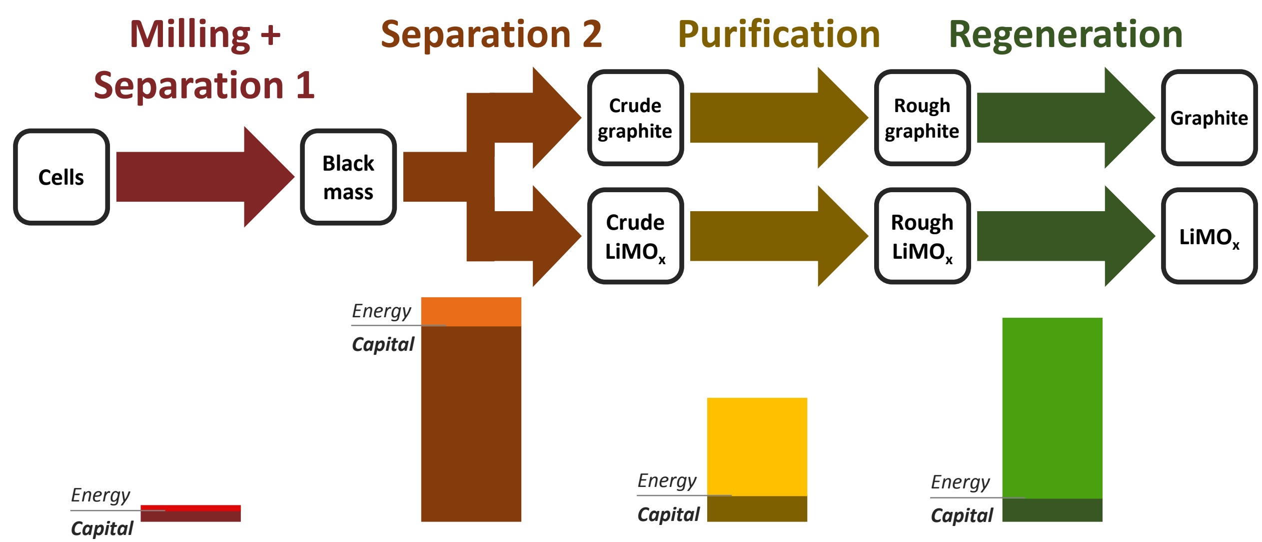
Technical Accomplishments – Technoeconomic Modeling

- A process model for direct recycling has been created.
- The model will be refined and integrated with manufacturing models and cell testing performance data to optimize value of active material recovery processes.
- Model is being extended for feedstock specific process modifications.



Equipment

Milling + Separation 1	Separation 2	Purification	Regeneration
Shredder	Reactor	Reactors	Mixer
Shaker Table	Dense Media	Centrifuge	Furnace
Magnetic Separator	Centrifuge	Furnace	

Example:

907 kg cells →	402 kg black mass →	129 kg Graphite →	92 kg Graphite →	73 kg Graphite
		212 kg $LiMO_x$ →	151 kg $LiMO_x$ →	120 kg $LiMO_x$

+ 80 kg Cu & 35 kg Al

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Responses to Reviewers' Comments

- **New Project**

Collaborations

- **Lawrence Berkeley National Laboratory (Robert Kostecki)**

  Advanced chemical diagnostics and materials characterization to guide recycling process development.

Challenges and Barriers

- **Feedstock complexity leading to unexpected chemical phenomena in the recycled active materials is the main technical challenge associated with this project.**

Proposed Future Research

- Extend direct recycling process to additional feedstocks – formed cells and complete modules
- Complete cell builds and testing to fully understand impact of recycled materials on technology lifetime
- Evaluate process compatibility with other cell chemistries
- Update technoeconomic model with materials performance and recycling process data; refine model for different feedstocks.

Summary

- A recycling process for direct recovery and reuse of Li-ion battery active materials has been scaled up and is being applied to multiple commercially-relevant feedstocks.
- Recycling will have a large impact on materials and costs associated with Li-ion technology.
- Initial work with “early” Li-ion chemistries; project will also evaluate process compatibility with newer cell chemistries.
- Cell builds with recycled active materials will provide data for quantitative technology valuation.